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THE EFFECTS OF GEOCHEMICAL PROCESSES ON THE TRANSPORT OF CONTAMINANTS IN MULTICOMPNENT SYSTEMS: A MODELING PERSPECTIVE

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### THE EFFECTS OF GEOCHEMICAL PROCESSES ON THE TRANSPORT

### OF CONTAMINANTS IN MUITICOMPONENT SYSTEMS: A MODELING PERSPECTIVE

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### ABSTRACT

An equilibrium geochemical transport model for multicomponent systems, TRANQL, was used to investigate the effects of geochemical processes on the transport of contaminants. TRANQL was used to investigate the seasitivity of cadmium transport to a range of initial conditions, pH, complexing ligand concentrations, and concentrations of a simultaneously sorbing solute. Aqueous-phase complexation, dissociation of water, and sorption were the processes considered. First, the transport of an initial pulse of cadmium with a constant concentration of EDTA was examined. Second, the transport of an initial pulse of cadmium and EDTA was simulated. The effects of variations in EDTA concentrations and pH were investigated. Finally, the transport of an initial pulse of cadmium and cobalt was examined. Results show a significant coupling between the geochemical processes of complexation and sorption and mass-transport. In cases where sorption is considered to be an important controlling reaction the transport of a solute in multicomponent systems is a strong function of the initial concentration and distribution of complexing ligands, pH, equilibrium formation constants, and the concentration of a simultaneously sorbing solute.

# INTRODUCTION

Geochemical transport modeling plays a significant role in characterising the macroscopic far-field environment and performance assessment of potential nuclear waste stor ge sites. The specific geochemical processes and the parameter values considered in the modeling affect the results of these transport simulations and thus the predicted performance of a repository. In particular most transport models currently used in nuclear waste storage programs rely solely on the use of a distribution coefficient to describe the equilibrium interaction and retardation of a radionuclide. However, distribution coefficient models do not take into account complex geochemical or sultisolute reactions and have often been inadequate in simulating observed transport. Implicit in these models is also the assumption that the solutes being modeled act independently of the bulk solution composition, i.e., the transport of one particular contaminant does not depend upon the presence of any other constituent in the system. Therefore, more sophisticated multicomponent equilibrium geochemical transport models have recently been developed (Valocchi et al., 1981; Charbeneau, 1981; Cederberg et al., 1985). Multicomponent models have the potential to incorporate the equilibrium chemical reactions which may be significant in describing and predicting transport such as aqueous-phase complexation, ion-exchange, simultaneous adsorption, dissociation of water, and precipitation/ dissolution. Because contamination sources and environments at potential nuclear waste repository sites are in reality multicomponent solutions, the application of multicomponent models to such sites should be investigated. As an initial step, one such model, TRANQL (Cederberg et al., 1985), was used to investigate the effects of geochemical processes on the transport of a sorbing solute, cadmium, in a multicomponent system.

The principal objective of this paper is to attempt to answer the question, "does including complex chemistry in transport models make a difference?", i.e., what is the impact of various geochemical processes and parameter values on the transport of a solute in a multicomponent system. In the sections that follow, first, the geochemistry considered in the simulations is described. Second, a general outline of the model, TRANQL, is given. Finally, the effects of changes in initial conditions, complexing ligand concentration, pH, and simultaneously sorbing solute concentrations are detailed. Aqueous-phase complexation, dissociation of water, and sorption were the geochemical processes considered. From these examples it can be concluded that a significant coupling between the geochemical processes of complexation and sorption and mass-transport exists. Further investigations should continue in applying this model to radionuclide transport at potential nuclear waste repository sites in order to validate or invalidate the use of distribution coefficient transport acdels.

### MODEL DESCRIPTION

TRANQL is the model used for the simulations in this paper (Cederberg et al., 1985). In TRANQL the equilibrium interaction chemistry is posed independently of the mass transport equations, which leads to a set of algebraic equations for the chemistry coupled to a set of differential equations for the mass transport. A solution is found by iterating between the two equation sets. The mass transport equations are solved using the Galerkin-Finite Element Method with an implicit time-stepping scheme and the algebraic chemical equation set is solved using the Newton-Raphson Method.

The equilibrium interaction chemistry considered here as a model system and shown in Table 1 includes scrption, complexation, and dissociation of water. The partitioning of species hased on these chemical reactions depends

upon a variety of physical and chemical conditions such as temperature, pH, the bulk solution composition, the type and number of sorption sites, and the respective concentration of each species in the system. The set of components which combine to form all species is defined as (Cd<sup>2+</sup>, Ca<sup>2+</sup>, HCO<sub>3</sub>, EDTA<sup>4-</sup>, SOH, H<sup>4</sup>) where SOH is the total number of sorption sites in moles/liter.

The value of the binding coefficient,  $K_{18}$ , for the sorption reaction of cadmium is a function of pH. Thus, two values of  $\log_{10}K_{18}$  are given in Table 1. The values of  $K_{18}$  were calculated from data taken from laboratory batch experiments (Leckie et al., 1983). The aquifer material used in the experiments came from a calcareous, sandy, unconfined aquifer near Borden, Ontario, Canada.

To solve the set of chemical equilibrium equations for the eighteen species a computer code, MICROQL (Westall, 1979), is used. The information required from MICROQL for input into the transport equations after the concentrations of all species are calculated is the total aqueous concentrations of all components. For example,  $\operatorname{Cd}_{AO}$  (the total amount of cadmium in the aqueous phase) =  $\operatorname{Cd}^{2-1}+\operatorname{CdEDTA}^{2-1}+\operatorname{CdHEDTA}^{-1}+\operatorname{CdHED$ 

For one-dimensional flow in a homogeneous, isotropic porous medium the fundamental transport equations in TRANQL are the following:

$$\frac{\delta(C_T)}{\delta t} = D_L \frac{\delta^2(C_{AQ})_i}{\delta x^2} - v \frac{\delta(C_{AQ})_i}{\delta x} \qquad i=1...N_c \quad (1)$$

where N = number of components (=6), (C<sub>T</sub>) = total (aqueous + sorbed phase) concentration of component i, (C<sub>AQ</sub>) = aqueous phase concentration of component i. (D<sub>L</sub> = coefficient of hydrodynamic dispersion, and v = average groundwater velocity. Instead of formulating the transport equations around the mass balance for each species in the aqueous phase, the transport equations have been written around the mass balance for the total concentration of each component. This formulation allows the interaction chemistry to be posed independently of the mass transport equations and coupling of the two sets in a precise manner. The time derivative of the total component concentration is a function only of the transport by advection and dispersion of the total aqueous component concentration. The (C<sub>AQ</sub>) s are determined from the solution of MICROQL, the equilibrium chemistry equation set.

The final set of mass-transport equations is a system of differential equations (in this example two equations). The final set of equilibrium chemistry equations is a system of algebraic equations (here eighteen equations). After initializing the system to equilibrium, Eq. 1 can be solved directly for (EDTA $_{\pi}$ ) because (EDTA $_{\pi}$ )=(EDTA $_{\Lambda}$ 0). Solution to Eq. 1 for the nonconservative component, (Cd $_{\pi}$ 1), is found by iterating between the two equation sets until some prescribed convergence tolerance is met for

the nonconservative corponent. It is assumed that that concentration of the other components, Ca , HCO , SOH, and H , remain constant.

	Reactions .		Species	log <sub>10</sub> K		
1	C4 2+	•	Cd <sup>2+</sup>	<u>_</u>		
2	Ca <sup>2+</sup>	•	Ca <sup>2+</sup>	0		
3	10C03	•	BC03	0		
4	EDTA <sup>4-</sup>	•	EDTA <sup>4-</sup>	0		
5	SOH	-	BOH	0		
6	<b>1</b> *	-	E+	0		
Complexation						
7	3E+ EDTA4-	-	ByEDTA"	6.16		
8	4E+ EDTA4-		<b>E</b> ARDTA	10.26		
9	Cd <sup>2+</sup> + EDTA <sup>4-</sup>			15.78		
10	$cd^{2+} + E^+ + EDTA^{4-}$			18.50		
11	Cd <sup>2+</sup> + BCO <sub>3</sub>			1.92		
12	Ua <sup>2+</sup> + EDTA <sup>4-</sup>			10.52		
13	$Ca^{2+} + H^+ + EDTA^{4-}$			13.00		
14	Ca <sup>2+</sup> + ECO <sub>3</sub>			0.92		
15	Cd <sup>2+</sup> - H <sup>+</sup> + H <sub>2</sub> O			-10.28		
16	Ca <sup>2+</sup> - H <sup>+</sup> + H <sub>2</sub> O	-	CaOH+	-12.78		
D180	ociation of Water					
17	-H+ H20	-	Off_	13.91		
Вогр	tion					
18	Cd <sup>2+</sup> + SOH	•	C48OH	5.01 pR = 7.46 5.35 pH = 7.97		

Table 1. Chemical Reactions for the cadmium/EDTA system.

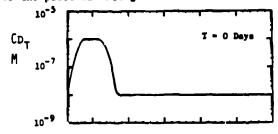
# EFFECTS OF GEOCHEMICAL PROCESSES

TRANQL was used to investigate the sensitivity of cadmium transport to a range of initial conditions, complexing ligand concentrations, pH, and concentrations of a simultaneously sorbing solute. For all the simulations in this paper the multicomponent geochemical parameter values and the physical parameter values are the same and held constant. The geochemical system considered is described in Table 1. Flow was in one dimension with  $\nu=0.10$  m/day and  $D_{\rm L}=0.006$  m/day.

### Effect of a Constant Initial EDTA Concentration

The effect of three different constant EDTA concentrations on the transport of an initial pulse of cadmium was\_investigated. The total concentrations of Ca , HCO , SOH, and pH are held constant throughout the simulations in order to isolate the effect of the constant EPTA concentration on the transport of cadmium. At time t = 0, a pulse of Cd is placed in the system which is then assumed to be at chemical equilibrium. Three different constant concentrations of EDTA (10 , 10 , and 2.51x10 M) were used in the simulations. The three respective initial Cd distributions are given in Figure 1. The boundary values are taken to be those at the extremes of the initial distributions. For numerical purposes the background level of Cd was assumed to be two orders of magnitude smaller

than the peak concentration value of the input pulse. Constant aqueous background concentrations for all components were used as the boundary conditions at x=-1.375 m. The center of the pulse was assigned x=0 at t=0.



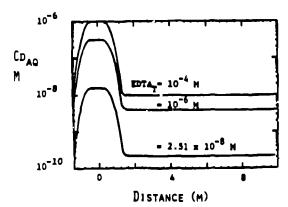


Figure 1. Initial Cd/constant EDTA system.

For each initial concentration of EDTA a different  $\operatorname{Cd}_{AQ}$  concentration exists. In Figure 1 it shows the more EDTA in the system the more cadmium complexes with the EDTA and remains in the aqueous phase. A two-order-of-magnitude decrease in EDTA concentration results in a two-order-of-magnitude decrease in  $\operatorname{Cd}_{AQ}$ . So while the initial concentration distribution of total cadmium,  $\operatorname{Cd}_{T}$ , is the same in all three cases, the  $\operatorname{Cd}_{AQ}$  distribution is quite different. The difference  $\operatorname{Cl}_{T}$  due to the amount of aqueous cadmium complexation which in turn is based on the chemical reactions and formation constants listed in Table 1.

TRANQL was run to simulate a real time of 40 days. A time step of 2 hr and nodal spacing of 0.25 m were used in the simulations. Figure 2 shows the results of the three TRANQL simulations, each with a different initial concentration of EDTA. The Lotal concentration of aqueous cadmium, Cd AO, is plotted as a function of distance for each EDTA concentration. The differences in the initial distributions of Cd resulted in marked differences in the results of the transport simulations. When EDTA is at its maximum value more cadmium complexes with the EDTA, romains in the aqueous phase, and therefore is transported further in the x-direction. When EDTA is at its minimum value. only 2% of the cadmium remains in the aqueous phose, 98% of the cadmium is immediately sorbed and virtually no cadmium is transported in the w-direction.

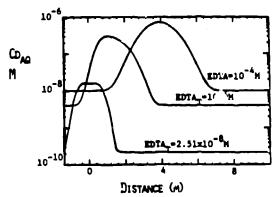


Figure 2. The effect of constant EDTA on Cd AO.

### Effect of an Initial Pulse of EDTA

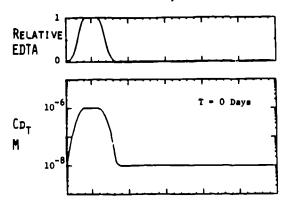
Instead of a constant concentration of EDTA a pulse of Cd<sub>m</sub> and EDTA<sub>m</sub> is placed in the system at time t = 0. Four different concentrations of EDTA in the input pulse were used. To reference the simulations back to the varying initial concentrations of EDTA a notation which specifies each case is listed in Table 2. Cases EDTA(la)-EDTA(4a) were used in this section to simulate the transport of a pulse containing EDTA and Cd. Cases EDTA(lb)-EDTA(4b) were used in the next section to investigate the effect of pH on the transport of Cd. The initial distribution of Cd<sub>a</sub> for Cases EDTA(la)-(4a) is shown in Figure 3. Initially, with more EDTA in the system more Cd<sub>a</sub> also exits. A two-order-cf-magnitude difference in peak concentration of EDTA results in a 50% decrease in the initial peak Cd<sub>aO</sub> concentration.

Case	Peak(H)	1	Background(M)	pН
EDTA(la)	2,51×10 <sup>-6</sup>	_	2.51×10 <sup>-8</sup> 5.01×10 <sub>-8</sub> 1.6×10 <sub>-6</sub> 1.0×10	7.97
EDTA(2a)	2.51x10 <sup>-6</sup> 5.01x10 <sub>4</sub>	7	5.01×10 <sup>-8</sup>	7.97
EDTA(3a)	1.0x10	7	1.Gx10 /	7.97
EDTA(44)	1.0x10 <sup>-3</sup> 1.0x10 <sup>-4</sup>	1	1.0x10 <sup>-0</sup>	7.97
EDTA(1b)	2.51×10 <sup>-6</sup> 5.01×10 <sup>-6</sup> 1.0×10 <sup>-6</sup>	1	2.51×10 <sup>-8</sup> 5.01×10 <sup>-8</sup> 1.0×10 <sup>-7</sup>	7.46
EDTA(2b)	5.01×10-6	1	5.01x10_8	7.46
EDTA(3b)	1.0x10 <sup>-3</sup>	1	1.0x10 <sup>-</sup> /	7.46
EDTA(4b)	1.0x10 <sup>-4</sup>	1	1.0x10 <sup>-6</sup>	7.46

Table 2. EDTA Concentrations used in cadmium/ EDTA simulations

Pigure 4 shows the results of four TRANQL simulations, each with a different initial pulse concentration of EDTA. When the peak concentration of EDTA is a maximum [Case EDTA(4a)], most of the cadmium is complexed with the EDTA and therefore remains in the aqueous phase. In this case ordnium be 'aves in a manner similar to that of EDTA, the conservative component. As the initial peak EDTA concentration decreases, more and more cadmium adsorbs to the porous medium due to the decrease in cadmium complexation. A two-order-of-magnitude decrease in peak EDTA

concentration [Case EDTA(4a) to EDTA(1a)] results in more than an order-of-magnitude decrease in the maximum coserved concentration of cadmium. As the peak EDTA concentration decreases, cadmium becomes more and more immobile and the concentration distribution becomes asymmetric.



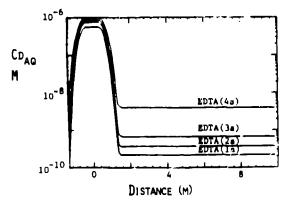


Figure 3. Initial Cd/pulse EDTA system.

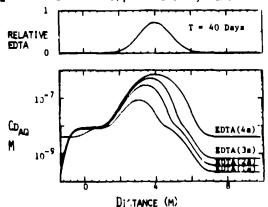


Figure 4. The effect of pulse EDTA on Cd AO.

The asymmetry in the cadmium distribution can be attributed to the changing speciation everywhere in the system over time and the initial pulse distribution of EDTA. The relative broadening and asymmetry of the Cd AD curves was not seen in the simulations where EDTA was constant. The asymmetry has been shown to result from the initial pulse distribution of EDTA and Cd and the ever-changing degree of geochemical

complexation, speciation, and sorption occurring everywhere in the system (Cederberg, 1985).

#### The Effect of pH

For the cadmium/EDTA system (see Table 1) the formation or binding coefficient, K18, for the sorption reaction is an implicit function of pH. As the pK increases, so does the value of the binding coefficient. Therefore, cases EDTA(1b)-EDTA(4b) were simulated to investigate the effect of pH on cadmium transport. For direct comparison the concentration of Cd AO for Cases EDTA(1b) and (1a) and EDTA(4b) and (4a) are shown in Figure 5 for t = 40 days. When the pH is lower the caimium is more mobile and transported further. The effect is most noticeable when the peak EDTA concentration is lowest [Cases EDTA(1b) and (1a)]. In this case the decrease in pH results in a 75% increase in the observed maximum cadmium concentration. The complexation of the cadmium with the EDTA overwhelms any change in cadmium concentration due to a change in pH. There is little noticeable change between the cadmium concentrations when the EDTA posk concentration is a maximum [Cases EDTA(4a)-(4b)]. Because the binding coefficient has a lower value at a lower pH, the cadmium will bind less strongly with the porous medium and the EDTA will compete for the cadmium more strongly.

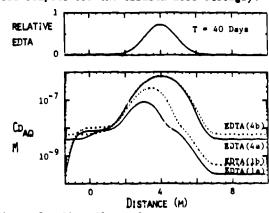


Figure 5. The effect of pH on Cd AO.

# The Effect of Cobalt

More than one reactive component is often found in contaminant systems. A second component may or may not compete with the first component for specific complexing ligands or sorption sites. To investigate the effects of the presence of a second sorbing component TRANQL was used to mimulate the transport of cadmium and cobalt. The additional chemical reactions to the cadmium/cobalt/EDTA system are listed in Table 3. The complete system of reactions includes all those reactions from Table 1 plus those reactions specific to cobalt. In this set of reactions cobait forms complexes similar to those of cadmium. The values of the binding coe, ficients for cadmium and cobalt in a system together were found by batch experiments.

The transport aquation set for the cadmium/cobalt/EDTA system contains one conservative transport equation for EDTA and two nonconservative transport equations, one each for the components, cadmium and cobalt. The physical parameter values used in the simulations are the same as those of the previous section. The initial and boundary conditions are also similar in value; however, here  $\mathrm{pH}=7.46$  and  $\mathrm{SOH_T}=0.204$  M. For this investigation a pulse of cadmium, cobalt, and EDTA is input while the concentrations of the other components are held constant and remain at the values described in the previous sections.

	Reactions	Species	108 <sub>10</sub> K
19	co <sup>2+</sup> =	Cc <sup>2+</sup>	0
20	Co <sup>2+</sup> + EDTA <sup>4-</sup> =	CoEDTA	15.58
21	$Co^{2+} + H^{+} + EDTA^{4-} =$	CoHEDTA	A 18.40
22	$co^{2+} + Hco_{3}^{-} =$	CoHCO3	2.72
23	Co <sup>2+</sup> - H <sup>+</sup> + H <sub>2</sub> O =	CoOH	-9.88
Sorpti 24	Co <sup>2+</sup> + SOH =	CoSOH	pH=7.46 3.0
(18)	Cd <sup>2+</sup> + SOH =	Са ѕон	3.6

Table 3. Additional chemical reactions for the cadmium/cobalt/EDTA system.

Simulations were run with two different peak concentrations of total cobalt (Co<sub>T</sub> = 10<sup>-3</sup> M and = 10<sup>-4</sup> M) present. Figure 6 shows the initial distributions of EDTA<sub>T</sub>, Cd<sub>T</sub>, and Co<sub>T</sub>. Case Col (represented by the dotted line) represents the simulation where Co<sub>T</sub> = 10<sup>-4</sup> M. Case Co2 (represented by the solid line) represents the simulations where Co<sub>T</sub> = 10<sup>-5</sup> M. Figure 7 shows the initial distributions of Cd<sub>T</sub> and Co<sub>T</sub> after chemical equilibrium have been attained. Because the Cd and Co compete for EDTA and sorption sites, the presence of a lower initial concentration of Co<sub>T</sub> (Case Col) results in a higher concentration of Cd<sub>T</sub> after the system comes to equilibrium. As the cobalt concentration decreases, less cobalt complexes with EDTA and more cadmium complexes with the EDTA. Thus, less cadmium is available to sorb and the Cd<sub>T</sub> increases. From these initial conditions one would expect Cd to be transported further and at a higher concentration for the case where Co<sub>T</sub> = 10<sup>-7</sup> M.

Figure 8 shows the results of two TRANQL simulations where the initial peak values of EDTA, is  $10^{-1}$  H and Cd, is  $10^{-1}$  H. As the concentration of cobalt increases, the concentration of Cd, decreases. The difference in Co concentration has a significant effect on the transport of cadmium. Although Cd, initially is higher when Co, is lower, the difference in the two Cd, distributions increases with time. Initially there was approximately a 40% difference in the Cd, peak value. At the end of 40 days an order-of-mignitude difference in the peak concentration of Cd, exists.

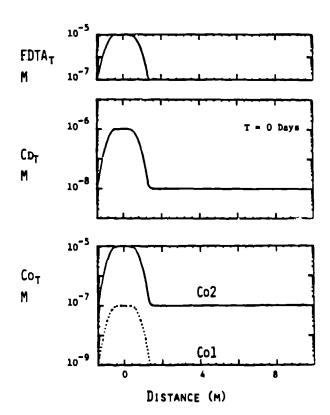


Figure 6. Initial Cd / Co EDTA system.

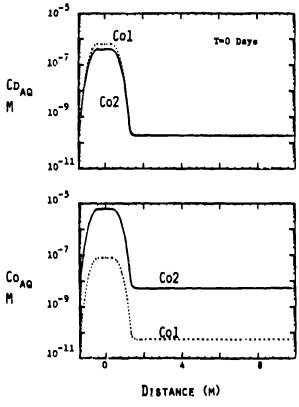


Figure 7. Initial Cd AQ CO AQ EDTA system.

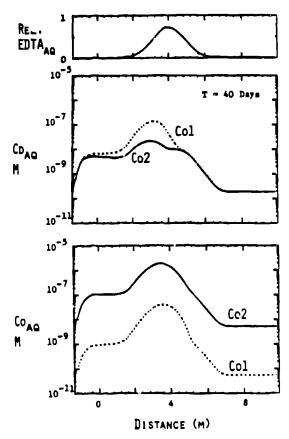


Figure 8. The effect of Co AO on Cd AO.

### SUMMARY

Results show a significant coupling between the geochemical processes of complexation and sorption and mass-transport. In cases where sorption is considered to be an important controlling reaction the transport of a solute in multicomponent systems is a strong function of the initial concentration and distribution of complexing ligands (here EDTA), equilibrium formation constants, pH, and the concentration of a simultaneously sorbing solute. In the presence of a conservative complexing component such as EDTA the mobility or transport of cadmium increased significantly as the concentration of EDTA incressed. A two-order-of-megnitude increase in EDTA resulted in an order-of-magnitude increase in the predicted maximum concentration of aqueous cadmium. When EDTA was constant, the Cinc curves retained their symmetry. However, in cases where a pulse of EDTA was input the Cd distributions became asymmetric and significantly broadened. Thus, the initial distributions and concentrations of all components in the system may effect the transport of a sorbing solute. As the pH decreased and the corresponding binding coefficient decreased, the transport of cadmium increased. When the EDTA peak concentration was a minimum, the decrease in pH resulted in a 75% increase in the predicted maximum Cd concentration. However, when the peak concentrate tion of EDTA was a maximum, the change in pH had little effect on cadeium transport. Finally, as the concentration of a second sorbing component ,cobalt, was increased, the transport of cadeium respectively decreased. A two-order-of-magnitude increase in cobalt resulted in a 50% decrease in the Cd AQ concentration.

The specific results from these simulations are a function of the set of reactions considered, the components present, and the equilibrium formation coefficients. However, the results do represent trends one might observe in ground-water contamination systems which concern the transport of inorganics in a multicomponent system. From the viewpoint of nuclear waste disposal programs the results of these calculations raise several important questions. First, what is the effect of multicomponent complexation and sorption on mass-transport and thus performance assessment at potential nuclear waste repository sites? Second if models such as TRANQL are needed are the necessary data being gathered in order to use these models? Finally, how valid is the application of distribution coefficient models to predict transport at repository sites? Further investigations are continuing in order to answer these questions.

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